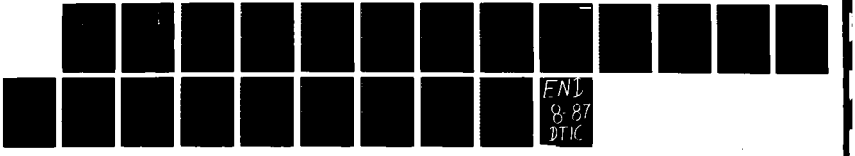
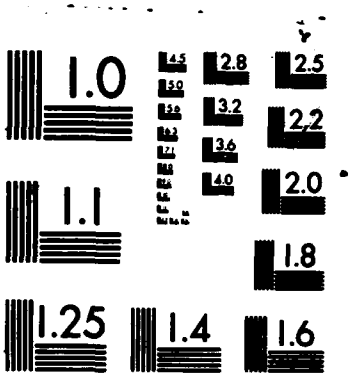
AD-A182 403 ADVANCED LASER SOURCE RESEARCH(U) STANFORD UNIV CA DEPT OF PHYSICS A L SCHAWLOW ET AL. 01 JUN 87 N00014-78-C-0403 1/1

UNCLASSIFIED F/G 9/3 NL



FNL
8-87
DTIC



(12)

* FINAL TECHNICAL REPORT *

U.S. Office of Naval Research

CONTRACT: N00014-78-C-0403

ADVANCED LASER SOURCE RESEARCH

CONTENTS

• • •

Arthur L. Schawlow

J.G.Jackson - C.J.Wood
Professor of Physics
Stanford University
Stanford, CA 94305-4060

June 1, 1987

^ FINAL REPORT ^

ADVANCED LASER SOURCE RESEARCH
U.S. OFFICE OF NAVAL RESEARCH

CONTRACT: N00014-78-C-0403

Principal Investigator: Professor Arthur L. Schawlow,
J.G.Jackson - C.J.Wood Professor of Physics

Co-Principal Investigators: Professor Theo W. Hansch,
Professor of Physics (on leave)
Professor Stephen E. Harris,
Professor of Electrical Engineering and Applied Physics
Professor Richard N. Zare,
Professor of Chemistry

NONLINEAR SPECTROSCOPY

by
Professor Arthur L. Schawlow

The approach has been to study molecular systems which exhibit unusual spectroscopic behavior, in the hope that they may lead to novel techniques for upconverting laser frequencies. We have also studies novel methods of laser spectroscopy and wavelength stabilization.

1. Two-Photon Transitions in Molecules

Previous work in this area has been summarized in our Annual Progress Reports. The general aim has been to understand the strongly enhanced two-photon transitions observed in diatomic alkali molecules, in order to explore their usefulness for lasers. These transitions, can have strengths nearly as great as for allowed single-photon transitions when

there is a very near coincidence of the laser frequency with an allowed transition to an enhancing level. Thus one can use visible light to obtain large numbers of highly excited states of the molecules. However, because of their symmetry, molecules excited to these states cannot make direct transitions to the ground electronic level in homonuclear molecules such as Na_2. We have, therefore, studied the mixed alkali vapor of Na and K, and have detected two types of two-photon transitions. Using a continuous-wave narrowband dye laser tunable from 570 to 584 nanometers, two-photon transitions in NaK free of Doppler broadening have been observed. Five Doppler-broadened transitions were also detected, and are ascribed to two-photon hybrid resonances involving the photolysis of K_2 and a subsequent transition in the excited K atom.

We have also used spectroscopy of fluorescence, resulting from two-photon excitation, as an aid in identification of two-photon transitions in Na_2. Several methods were introduced and used to identify the transitions and analyze the spectra. Using these methods, the upper level rotational quantum number, energy and electronic character could be determined without any assumptions about the state to which the upper level belongs. Twenty-nine two-photon transitions were observed and analyzed. Of these, 19 could be fitted to the Dunham rotation-vibration constants of the $^1\Sigma_g^+$ (3s + 5s) state. Since the wavelengths of these new transitions were measured more accurately than those in previous studies using pulsed lasers, improved values of the Dunham coefficients for that state could be derived. They were in good agreement with the values obtained from the best polarization labeling experiments.

2. High-Contrast Saturation Spectroscopy for Optically Thick Samples

Doppler-free saturation spectroscopy in the regime of strong pumping intensities and optically thick atomic samples was investigated experimentally and theoretically. It was shown that a high signal-to-background ratio can be obtained and, at the same time, subnatural linewidths can be reached. It was also shown that further contrast and linewidth improvements can be obtained by supplementing the primary depleted pumping beam with a second pumping beam. By using the signal beam from a first setup as the pumping beam for a second identical arrangement, extreme values for contrast and linewidth should be obtainable.

FINAL REPORT

Advanced Laser Source Research
U.S. Office of Naval Research
Contract N00014-78-C-0403

Tunable Lasers and Coherent Light Techniques for High Resolution Ultraviolet Spectroscopy -

ed → (to ! 11)

by

Professor Theo W. Hansch

In May 1986, T. W. Hansch left Stanford University in order to accept a new appointment as Director at the Max-Planck Institute for Quantum Optics in Garching, Germany, and as Professor at the University of Munich.

His research program in the Department of Physics at Stanford University has since been carried on by three graduate students and two visiting scientists:

Raymond G. Beausoleil, received Ph.D. Winter, 1987
David H. McIntyre, to receive the Ph.D., June, 1987
Gregory M. Cutler, to receive the Ph.D., June, 1987
Dr. Bernard Couillaud, Coherent, Inc., Palo Alto, CA
Dr. Christopher J. Foot, Clarendon Laboratory, Oxford, England

The following papers acknowledge ONR support, and have been published or submitted since January 1986:

PHYSICAL REVIEW A VOLUME 33, NUMBER 3 MARCH 1986

Ultrahigh-resolution two-photon optical Ramsey spectroscopy of an atomic fountain

R. G. Beausoleil and T. W. Hänsch
Department of Physics, Stanford University, Stanford, California 94305-2196
(Received 19 September 1985)

We present a semiclassical analysis of ultrahigh-resolution two-photon optical Ramsey spectroscopy of cold neutral atoms falling freely in a fountain. Considering atoms which interact with the same standing-wave laser field twice on their parabolic trajectories and averaging over a broad atomic velocity distribution, we predict a nearly Lorentzian line shape whose width is just the natural linewidth. We have investigated a number of systematic corrections to the atomic resonance frequency, including first-order and second-order Doppler shifts, gravitational red shifts, and ac Stark shifts. First-order Doppler shifts due to vertical motion cancel even if the counterpropagating beams are slightly misaligned, and resolutions below 1 Hz appear feasible with a fountain of modest dimensions.

Reprinted from Optics Letters, Vol. 11, page 97, February 1986

Continuous-wave second-harmonic generation as a surface microprobe

G. T. Boyd and Y. R. Shen
Department of Physics, University of California, Berkeley, California 94720

T. W. Hänsch
Department of Physics, Stanford University, Stanford, California 94305

Received October 22, 1985, accepted November 26, 1985

We demonstrate that monolayer detection by optical second-harmonic generation is possible even with a 20-mW cw diode laser. Using a focused scanning laser beam, we also demonstrate a new method of monolayer surface microscopy. The same experiments suggest the possibility of an optical monolayer memory device.

VOLUME 56, NUMBER 6 PHYSICAL REVIEW LETTERS 10 FEBRUARY 1986

Measurement of the 1*S*-2*S* Frequency in Atomic Hydrogen

E. A. Hildum, U. Boesl,[a] D. H. McIntyre, R. G. Beausoleil, and T. W. Hänsch
Department of Physics, Stanford University, Stanford, California 94305
(Received 24 October 1985)

We report on a first precise measurement of the 1*S*-2*S* energy interval in atomic hydrogen. Observing the 1*S*-2*S* transition in an atomic beam by pulsed Doppler-free two-photon spectroscopy and using an interferometrically calibrated absorption line of $^{130}Te_2$ at 486 nm as the reference, we measure the frequency $f(1S\text{-}2S) = 2\,466\,061\,395\,6(4.8)$ MHz. Using the calculated 1*S* Lamb shift, we obtain a value for the Rydberg constant, $R_\infty = 109\,737.314\,92(22)\ \text{cm}^{-1}$, which is not in good agreement with the most recent previous measurement.

HIGH RESOLUTION LASER SPECTROSCOPY OF ATOMIC HYDROGEN

R.G.Beausoleil, B.Couillaud, C.J.Foot, T.W.Hansch
E.A.Hildum, and D.H.McIntyre

Abstract

Although spectroscopy of hydrogen has played a central role in the development of atomic theory and quantum mechanics, the resolution of optical spectral lines remained limited by Doppler broadening to about one part in 10^3 until 1971. Since then, methods of Doppler-free laser spectroscopy have achieved major improvements. Thanks to recent experimental advances, the rate of progress has quickened dramatically in 1985, promising unprecedented opportunities for precision measurements of fundamental constants and for stringent tests of basic physics laws.

REFERENCE: Proceedings of the International Laser Science Conference, Dallas, Texas, November 18-22, 1985

THE HYDROGEN ATOM IN A NEW LIGHT

T.W.Hansch, R.G.Beausoleil, U.Boesl, B.Couillaud,
C.J.Foot, E.A.Hildum, and D.H.McIntyre

Abstract

It is well known that lasers and coherent light techniques have revolutionized high resolution spectrsocopy. Today we have at our disposal a powerful arsenal of techniques which can overcome the Doppler broadening of spectral lines, achieving ever higher spectral resolution. At Stanford, we have long been fascinated by the prospects of applying such tools to atomic hydrogen. As the simplest of the stable atoms, hydrogen permits unique confrontations between experiment and quantum electrodynamic theory. After briefly reviewing past spectroscopic studies of hydrogen, we will report on some recent experimental advances which have opened the door to dramatic future improvements in resolution, creating unprecedented opportunities for precision measurements of fundamental constants and for stringent tests of basic physics laws.

REFERENCE: in Methods of Laser Spectroscopy, edited by Y.Prior A.Ben-Reuven, and M.Rosenbluh (Plenum Publishing Corp., 1986)

HIGH RESOLUTION LASER SPECTROSCOPY OF ATOMIC HYDROGEN: ADVANCES AND PROSPECTS

T.W.Hansch, R.G.Beausoleil, B.Couillaud, E.A.Hildum, and D.H.McIntyre

Abstract

Recent advances in high resolution laser spectroscopy of the simple hydrogen atom promise exciting new opportunities for tests of fundamental physics laws and for precision measurements of the Rydberg constant, the photon/electron mass ratio, and the charge radii of the proton and deuteron.

REFERENCE: in Proceedings of the Workshop on Fundamental Muon Physics: Atoms, Nuclei, and Particles, Los Alamos, NM, January 20-22, 1986.

RAPID COMMUNICATIONS

PHYSICAL REVIEW A VOLUME 34, NUMBER 5 NOVEMBER 1986

Absolute calibration of the $^{130}Te_2$ reference line for positronium $1^3S_1-2^3S_1$ spectroscopy

D. H. McIntyre and T. W. Hänsch*

Department of Physics, Stanford University, Stanford, California 94305

(Received 18 July 1986)

We report on an interferometric frequency measurement of the $^{130}Te_2$ absorption line which has been used as a reference in Doppler-free two-photon spectroscopy of the positronium $1^3S_1-2^3S_1$ transition. The measurement relies on a comparison with a nearby, previously calibrated line of $^{130}Te_2$. The result, 616 803 545.98(71) MHz, lowers the observed value of the positronium $1^3S_1-2^3S_1$ interval to 1 233 607 142.9(10.7) and lessens the agreement between theory and experiment.

Optical Rydberg Transitions Following Electron Capture Into Slow, Highly Ionized Neon Recoil Ions

L.J. Lembo, Ch. Stoller, K. Danzmann, W.E. Meyerhof, and T.W. Hänsch[a]

Department of Physics, Stanford University, Stanford CA 94305

R. Gerson

Hamamatsu Corporation, Middlesex, N.J. 08846

Abstract

Emission spectra in the visible and near ultraviolet have been observed for transitions between Rydberg states of highly charged neon ions (Ne^{q+} with q=9 to 5), following electron capture from Na. Structure appears in those spectra for which the incoming ions carry L-shell core electrons. Together with data from polarization studies, these spectra reveal the influence exerted by core interactions in electron capture processes.

REFERENCE: Phys. Rev. A. (submitted for publication)

(Submitted to Physical Review A - Rapid Communications)

INTERFEROMETRIC FREQUENCY MEASUREMENT OF A $^{130}Te_2$ REFERENCE LINE FOR MUONIUM *1S-2S* SPECTROSCOPY

D. H. McIntyre and T. W. Hänsch*

Department of Physics, Stanford University

Stanford, CA 94305

ABSTRACT

We report on an interferometric frequency measurement of a $^{130}Te_2$ absorption line which can be used as a reference in Doppler-free two-photon spectroscopy of the muonium *1S-2S* transition. The frequency of this 488 nm $^{130}Te_2$ transition is determined with a precision of 8 parts in 10^{10} by comparison with a previously calibrated $^{130}Te_2$ line near 486 nm.

CONTINUOUS-WAVE MEASUREMENT OF THE *1S* LAMB SHIFT IN ATOMIC HYDROGEN

R. G. Beausoleil,[a] D. H. McIntyre, C. J. Foot,[b]

E. A. Hildum,[c] B. Couillaud,[d] and T. W. Hänsch[e]

Department of Physics, Stanford University

Stanford, CA 94305

ABSTRACT

We have determined the *1S* Lamb shift in atomic hydrogen with a precision of 2 parts in 10^4 by measuring the frequency of the *1S-2S* transition observed by continuous-wave Doppler-free two-photon spectroscopy. We employ an interferometrically calibrated absorption line in $^{130}Te_2$ as our reference and obtain a *1S-2S* interval of 2 466 061 413.8(1.5) MHz. Choosing a value of the Rydberg constant measured independently by high resolution spectroscopy of the hydrogen Balmer-β transition, we find the *1S* Lamb shift to be 8 173.3(1.7) MHz, in good agreement with the theoretical value of 8 172.94(9) MHz.

Final Report

For

The Office of Naval Research

For

Contract Number N00014-78-C-0403

Co-Principal Investigators:

S. E. Harris

J. F. Young

May 1987

Section 1

Introduction

This is the final report for the ONR contract spanning the period from 1 April 1978 to 31 March 1987. The overall theme of this contract has been on studies of the physics and technology of extreme ultraviolet (XUV) and soft x-ray lasers and devices. Much of the work has centered on methods for the production of core-excited metastable atoms and ions; and on the use of these atoms and ions both for new types of spectroscopy and for new types of XUV lasers.

This has been an extraordinarily productive period. Particular highlights of the work during this period include: The invention and demonstration of the anti-Stokes radiation source, the use of laser produced plasmas for creating high density of metastable species, and the proposals of both metastable transfer and Auger lasers.

In recent weeks we have demonstrated a 109 nm laser with an output of about 20μ J in a beam with 5 mRad divergence.

Section 2 of this report lists the highlights of the work during this contract period. Section 3 lists the names of students who have received their Ph.D. during this period. Section 4 gives a bibliography of publications which have resulted from this contract.

We note that much of this work has been jointly supported by contracts with the Army Research Office, the Air Force Office of Scientific Research, the Strategic Defense Initiative Organization, and the Lawrence Livermore National Laboratory.

Section 2

Summary of Principal Contributions

Highlights of our work in the areas of core-excited spectroscopy and lasers include:

1. The anti-Stokes radiation source was proposed and analyzed in the context of a two photon blackbody.

2. This radiation source was used to take the $3p$ absorption spectrum of neutral potassium at a resolution of about $2\,cm^{-1}$, which is the highest resolution reported for these features.

3. This source was also used to produce a 600 ps pulse of (incoherent) 56.9 nm radiation.

4. The class of quartet-doublet laser systems was proposed; the emission spectrum of neutral Li (near 20 nm) was taken, and the quartet and doublet manifolds of Li were experimentally connected for the first time.

5. First microwave, and later hollow-cathode, technology was used to produce metastable populations and to demonstrate several new types of core-excited spectroscopy. In one of these experiments the Grotrian diagram of core-excited Na was defined for the first time.

6. The laser-produced plasma method of creating high densities of core metastables was proposed and demonstrated.

7. A new dichroic beam splitter was invented by R. W. Falcone, and tested.

8. The method of core excitation by x-ray-produced electrons was proposed and demonstrated.

9. The ideas of shake-up and super-Auger lasers were suggested.

10. A novel longitudinal pumping geometry was suggested.

11. The quasi-metastable concept was suggested and verified in several experiments. Recently it has been used to form the basis of entirely new type of XUV spectroscopy, which provides an unprecedented capability for the measurement of XUV levels and autoionizing times.

12. We have demonstrated a 109 nm laser with an output power of about 20μ J in a 5 mrad divergence beam. The laser is based on an Auger process in neutral Xe and uses a novel type of traveling wave laser produced plasma excitation.

Section 3
List of Students

The students who have received their Ph.D. during the course of this period are listed below:

W. R. Green	March 1979
J. C. White	April 1979
J. H. Newton	September 1979
G. A. Zdasiuk	December 1980
M. D. Wright	March 1981
J. R. Willison	May 1982
J. E. Rothenberg	October 1982
J. C. Wang	March 1984
D. E. Holmgren	October 1984
K. D. Pedrotti	December 1985
A. J. Mendelsohn	May 1985
P. J. K. Wisoff	June 1986
D. P. Dimiduk	October 1986

Section 4

Bibliography

1. S. E. Harris, J. Lukasik, J. F. Young, and L. J. Zych, "Anti-Stokes Emission as a VUV and Soft X-Ray Source," in **Picosecond Phenomena**, C.V. Shank, E. P. Ippen, and S. L. Shapiro, eds. (New York: Springer-Verlag, 1978), pp. 76–84.

2. R. W. Falcone, J. R. Willison, J. F. Young, and S. E. Harris, "Measurement of the He $1s2s\,^1S_0$ Isotopic Shift Using a Tunable VUV Anti- Stokes Light Source," Opt. Lett. **3**, 162–163 (November 1978).

3. R. W. Falcone, "Inversion of Atoms and Molecules to the Ground State by Optical Pumping," Appl. Phys. Lett. **34**, 150–152 (January 1979).

4. S. E. Harris and J. F. Young, "Techniques for Rapid Laser Induced Energy Transfer Using Metastable States," in **Proceedings for the International Conference on Lasers '78**, Vincent J. Corcoran, ed. (McLean, Va.: STS Press, 1979), pp. 679–688.

5. W. R. Green, J. Lukasik, J. R. Willison, M. D. Wright, J. F. Young, and S. E. Harris, "Measurement of Large Cross Sections for Laser-Induced Collisions," Phys. Rev. Lett. **42**, 970–973 (April 1979).

6. J. C. White, G. A. Zdasiuk, J. F. Young, and S. E. Harris, "Observation of Atomic Pair Absorption With an Incoherent Source," Opt. Lett. **4**, 137–139 (May 1979).

7. W. R. Green, M. D. Wright, J. F. Young, and S. E. Harris, "Laser Induced Charge Transfer to an Excited Ionic State," Phys. Rev. Lett. **43**, 120–123 (July 1979).

8. W. R. Green, M. D. Wright, J. Lukasik, J. F. Young, and S. E. Harris, "Observation of a Laser-Induced Dipole-Quadrupole Collision," Opt. Lett. **4**, 265–267 (September 1979).

9. S. E. Harris, J. F. Young, W. R. Green, R. W. Falcone, J. Lukasik, J. C. White, J. R. Willison, M. D. Wright, and G. A. Zdasiuk, "Laser Induced Collisional and Radiative Energy Transfer," in **Laser Spectroscopy IV**, H. Walther and K. W. Rothe, eds. (New York: Springer-Verlag, 1979), pp. 349–359.

10. S. E. Harris, "Proposal for a 207 Å Laser in Lithium," Opt. Lett. **5**, 1–3 (January 1980).

11. J. R. Willison, R. W. Falcone, J. C. Wang, J. F. Young, and S. E. Harris, "Emission Spectra of Core-Excited Even Parity ^2P States of Neutral Lithium," Phys. Rev. Lett. **44**, 1125–1128 (April 1980).

12. Joshua E. Rothenberg and S. E. Harris, "XUV Lasers by Quartet to Doublet Energy Transfer in Alkali Atoms," IEEE J. Quant. Elect. QE-17, 418–422 (March 1981).

13. S. E. Harris, J. F. Young, R. W. Falcone, W. R. Green, D. B. Lidow, J. Lukasik, J. C. White, M. D. Wright, and G. A. Zdasiuk, "Laser Induced Collisional Energy Transfer," in Atomic Physics 7, D. Kleppner and F. M. Pipkin, eds. (New York: Plenum Press, 1981), pp. 407–428.

14. Joshua E. Rothenberg, J. F. Young, and S. E. Harris, "High-Resolution Extreme-Ultraviolet Spectroscopy of Potassium Using Anti-Stokes Radiation," Opt. Lett. 6, 363–365 (August 1981).

15. S. E. Harris, R. W. Falcone, M. Gross, R. Normandin, K. D. Pedrotti, J. E. Rothenberg, J. C. Wang, J. R. Willison, and J. F. Young, "Anti-Stokes Scattering as an XUV Radiation Source," in Laser Spectroscopy V, A. R. W. McKellar, T. Oka, and B. P. Stoicheff, eds. (New York: Springer- Verlag, 1981), pp. 437–445.

16. R. W. Falcone and K. D. Pedrotti, "Pulsed Hollow-Cathode Discharge for XUV Lasers and Radiation Sources," Opt. Lett. 7, 74–76 (February 1981).

17. J. R. Willison, R. W. Falcone, J. F. Young, and S. E. Harris, "Laser Spectroscopy of Metastable Extreme-Ultraviolet Levels in Lithium Atoms and Ions," Phys. Rev. Lett. 47, 1827–1829 (December 1981).

18. S. E. Harris, J. F. Young, R. W. Falcone, Joshua E. Rothenberg, J. R. Willison, and J. C. Wang, "Anti-Stokes Scattering as an XUV Radiation Source and Flashlamp," in Laser Techniques for Extreme Ultraviolet Spectroscopy, T. J. McIlrath and R. R. Freeman, eds. (New York: AIP, 1982), pp. 137–152.

19. R. W. Falcone, D. E. Holmgren, and K. D. Pedrotti, "Hollow-Cathode Discharge for XUV Lasers and Radiation Sources," in Laser Techniques for Extreme Ultraviolet Spectroscopy, T. J. McIlrath and R. R. Freeman, eds. (New York: AIP, 1982), pp. 287–295.

20. S. E. Harris, J. F. Young, R. W. Falcone, Joshua E. Rothenberg, and J. R. Willison, "Laser Techniques for Spectroscopy of Core-Excited Atomic Levels," in Atomic and Molecular Physics Close to Ionization Thresholds in High Fields, J. P. Connerade, J. C. Gay, and S. Liberman, eds. (France: Les Editions de Physique, 1982), pp. 243–254.

21. R. G. Caro, J. C. Wang, R. W. Falcone, J. F. Young, and S. E. Harris, "Soft X-Ray Pumping of Metastable Levels of Li^+," Appl. Phys. Lett. 42, 9–11 (January 1983).

22. R. W. Falcone and J. Bokor, "Dichroic Beamsplitter for Extreme Ultraviolet and Visible Radiation," Opt. Lett. 8, 21–23 (1983).

23. J. C. Wang, R. G. Caro, and S. E. Harris, "A Novel Short-Pulse Photoionization Electron Source: Li(1s2s2p) $^4P^0$ Deexcitation Measurements in a Plasma," Phys. Rev. Lett. **51**, 767-770 (August 1983).

24. S. E. Harris, J. F. Young, R. G. Caro, R. W. Falcone, D. E. Holmgren, D. J. Walker, J. C. Wang, Joshua E. Rothenberg, and J. R. Willison, "Laser Techniques for Extreme Ultraviolet Spectroscopy," in **Laser Spectroscopy VI**, H. P. Weber and W. Luthy, eds. (New York: Springer-Verlag, 1983), pp. 376-381.

25. D. E. Holmgren, R. W. Falcone, D. J. Walker, and S. E. Harris, "Measurement of Lithium and Sodium Metastable Quartet Atoms in a Hollow Cathode Discharge," Opt. Lett. **9**, 85-87 (March 1984).

26. R. G. Caro, J. C. Wang, J. F. Young, and S. E. Harris, "The Use of Laser-Produced Soft X-Rays for the Production of Excited Metastable Ions," Phys. Rev. A **30**, 1407-1418 (September 1984).

27. R. G. Caro, J. C. Wang, J. F. Young, and S. E. Harris, "The Excitation of Metastable Extreme Ultraviolet Levels," in **Laser Techniques in the Extreme Ultraviolet**, S. E. Harris and T. B. Lucatorto, eds. (New York: AIP, 1984), pp. 417-426.

28. P. J. K. Wisoff and R. G. Caro, "A Superheated Na Cell for X-Ray Photoionization Experiments," Appl. Phys. B **35**, 65-69 (1984).

29. S. E. Harris, R. G. Caro, R. W. Falcone, D. E. Holmgren, J. E. Rothenberg, D. J. Walker, J. C. Wang, J. R. Willison, and J. F. Young, "Metastability in the XUV: Lasers and Spectroscopy," in **Atomic Physics 9**, R. S. Van Dyck, Jr. and E. N. Fortson, eds. (Singapore: World Scientific Publishing Co., 1985), pp. 462-479.

30. R. G. Caro, J. C. Wang, J. F. Young, and S. E. Harris, "X-Ray Excitation of Energetic Metastable Levels in Atoms and Ions," Opt. News **11**, 9-11 (March 1985).

31. A. J. Mendelsohn and S. E. Harris, "Proposal for an Extreme Ultraviolet Selective Autoionization Laser in Zn III," Opt. Lett. **10**, 128-130 (March 1985).

32. P. J. K. Wisoff, R. G. Caro, and G. Mitchell, "A High Power Picosecond Dye Oscillator Synchronously Pumped by a Q-Switched, Mode-Locked Nd:YAG Laser," Opt. Commun. **54**, 353-357 (July 1985).

33. R. G. Caro and J. C. Wang, "Population Distributions in Li Vapor Excited by a Photoionization Electron Source," Phys. Rev. A **33**, 2563-2574 (April 1986).

34. P. J. K. Wisoff, R. G. Caro, and G. Mitchell, "High Power Picosecond Blue Pulses from a Dye Laser Synchronously Pumped by a Q-Switched, Mode-Locked Nd:YAG Laser," Appl. Optics **25**, 2250-2252 (July 1986).

35. D. J. Walker, R. G. Caro, and S. E. Harris, "Proposal for an Extreme Ultraviolet Auger Laser at 63.8 nm in Ce III," J. Opt. Soc. Am. B 3, 1515–1517 (November 1986).

36. R. G. Caro, P. J. K. Wisoff, G. Y. Yin, D. J. Walker, M. H. Sher, C. P. J. Barty, J. F. Young, and S. E. Harris, "Soft X-Ray Pumping of Inner Shell Excited Levels for Extreme Ultraviolet Lasers," in **Short Wavelength Coherent Radiation: Generation and Applications**, D. T. Attwood and J. Bokor, eds. (New York: AIP, 1986), pp. 145–156.

37. J. F. Young, J. J. Macklin, and S. E. Harris, "Ellipsoidal Focusing of Soft X-Rays for Longitudinally Pumping Short Wavelength Lasers," in **Short Wavelength Coherent Radiation: Generation and Applications**, D. T. Attwood and J. Bokor, eds. (New York: AIP, 1986), pp. 86–88.

38. J. F. Young, J. J. Macklin, and S. E. Harris, "Grazing-incidence ellipsoidal reflector for longitudinally pumping short-wavelength lasers," Opt. Lett. 12, 90–92 (February 1987).

39. S. E. Harris, and J. F. Young, "Core-Excited Metastable levels: Application to Spectroscopy, to the Generation of Picosecond XUV Pulses, and to Lasers," J. Opt. Soc. Am. B 4 547–562 (April 1987).

40. Guang-Yu Yin, C. P. J. Barty, D. A. King, D. J. Walker, S. E. Harris, and J. F. Young, "Low Energy Pumping of a 108.9 nm Xe Auger Laser," Opt. Lett. 12, 331–333 (May 1987)

GAS-SURFACE INTERACTIONS

Professor Richard N. Zare, Principal Investigator

Scattering Studies; Review of Research

Greg O. Sitz, Andrew C. Kummel,

Melissa A. Hines, and Hope A. Michelsen

We have completed an extensive study of the scattering of N_2 off the flat, close-packed Ag(111) surface. We measured not only the rotational population of the scattered N_2, but also the alignment as described by the quadrupole and hexadecapole moments of the $\vec{J}$ distribution, denoted by $A_0^{(2)}$ and $A_0^{(4)}$, respectively. We have performed several unique measurements in the field of gas surface interactions: this is the first time a polarization study has been performed on any molecule except NO; this is the first time an $A_0^{(4)}$ moment has been measured, and this is the first time the polarization has been recorded as a function of the incident gas energy or the surface temperature. In addition to the experimental work, we developed the theoretical framework required to extract the polarization of N_2 using 2+2 multiphoton ionization (MPI).

The experimental setup is conceptually quite simple. Nitrogen is expanded from a free jet in order to create a monoenergetic beam of N_2 in its lowest rotational states. This beam is directed at a Ag(111) surface which has been sputtered, annealed, and checked for cleanliness and crystallographic integrity using Auger and LEED spectroscopies. The energy of the beam can be varied either by heating the nozzle or seeding the N_2 in a

lighter gas, such as He or N_2. The silver surface is mounted on a manipulator which allows us to vary its temperature and geometric orientation with respect to the incident beam. The beam scatters off the surface and creates N_2 in high rotational states; the scattered beam is intercepted by the focus of a frequency-doubled YAG-pumped dye laser. The N_2 is state selectively ionized using 2+2 MPI and is detected using a time-of-flight mass spectrometer. In addition to scanning the dye laser wavelength to record the population of different rotational states, we can vary the direction of the linear polarization vector of the laser beam using a double Fresnel romb and record the ionization probability of a single rotational state as a function of the polarization of the laser. These two methods allow us to record both the population and alignment of the N_2 scattered off the silver surface.

Our first experimental finding is that the rotational distribution of the scattered N_2 is very similar to that of NO off of Ag(111) except for N_2 the high energy rainbow is much sharper at low energies. This is an important result because the rainbow features in the NO experiment have been attributed to a head-tail anisotropy in the NO-gas potential in which the N end is considered to be attractive and the O end repulsive; obviously, this is not a viable explanation for the N_2 data.

We also determined that the scattered nitrogen molecules in high rotational states are very polarized with their angular momentum vectors lying parallel to the plane of the silver surface: the molecules cartwheel (tumble) across the surface. In our first experiments, we determined the polarization of N_2 by comparing the intensities of two different rotational branches. This method works quite well when the molecules are 100%

polarized because with this assumption it is reasonable to assume a relationship between $A_0^{(2)}$ and $A_0^{(4)}$. In our most recent experiments we determined the alignment by directly varying the polarization of the laser. The allows us to determine accurately the polarization of the lower rotational states which are only slightly aligned. Our findings are quite fascinating; the lowest rotational states have their angular momentum vectors pointing toward the surface normal: the molecules spin on the surface. The highest rotational states have a delta function for their distribution of angular momentum vectors: they are 100% polarized and cartwheel across the surface. We could only determine that this distribution was a delta function by independently measuring $A_0^{(2)}$ and $A_0^{(4)}$ using our new technique.

The interpretation is a simple one: for a flat rigid surface, the forces are directed along the surface normal, causing a torque to be exerted on the anisotropic N_2 projectile so that it rotates with its angular momentum at right angles to this force, i.e., with its angular momentum lying in the plane of the flat surface. The surface scattering process acts as an M state selector!

We determined the polarizations and rotational populations as a function of beam energy and surface temperature. While the surface temperature did not significantly affect the scattered distribution, the beam energy had two pronouced effects. Increasing the beam energy, raises the quantum number of the high energy rotational rainbow and increases the number of rotational states which are 100% polarized.

Presently, we are trying a completely new technique to measure the in-plane forces which the scattered molecules experience. If there are corru-

gations or frictional forces, the scattered nitrogen molecules should have a net helicity (spin), i.e., they will tend to rotate clockwise versus counterclockwise. When a football is thrown at high speed and lands on a grass field, it almost always tumbles in the direction thrown; we want to determine if an analogous mechanism is operating in our system. If this experiment is successful (our recent results are quite encouraging), it will yield a profound insight into how energy is transferred between gases and surface electrons via "frictional forces." As far as we know, theoreticians have always neglected this effect, and this may be a serious oversight. We have already developed the theory required to analyze these measurements of molecular orientation (net helicity) using 2+2 MPI.

END

8-87

DTIC